This Page Is Inserted by IFW Operations and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problem Mailbox.

THIS PAGE BLANK (USPTO)

(19) World Intellectual Property Organization International Bureau





(43) International Publication Date 10 May 2001 (10.05.2001)

PCT

(10) International Publication Number WO 01/32735 A1

- (51) International Patent Classification⁷: C08G 18/48, 18/10
- (21) International Application Number: PCT/EP00/09731
- (22) International Filing Date: 5 October 2000 (05.10.2000)
- (25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

99121697.9 2 November 1999 (02.11.1999)

- (71) Applicant (for all designated States except US): HUNTS-MAN INTERNATIONAL LLC [US/US]; 500 Huntsman Way, Salt Lake City, UT 84108 (US).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): PARFONDRY, Alain [FR/BE]; Rue Longue 82, B-1150 Brussels (BE). YU, Jianming [CN/BE]; Avenue Charles Woeste 66, B-1090 Brussels (BE).

- (74) Agents: BAKEN, Philippus, Johannes, Leonardus, Henricus et al.; Huntsman ICI (Europe) BVBA, Huntsman Polyurethanes, Intellectual Property Department, Everslaan 45, B-3078 Everberg (BE).
- (81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

- With international search report.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

10

15

20

25

30

35

PROCESS FOR MAKING HIGH RESILIENCE FOAMS

The instant invention relates to a process for making high resilience foams, and to reaction systems that are useful in the said process.

EP-A-0547765 to Bleys et al. discloses a process for making high resilience foams comprising reacting high 4,4' MDI with an EO-rich polyol. While the foams thus obtained exhibit good properties, the process suffers from two main drawbacks. The first drawback is that this process, if to be used on large scale, necessarily makes use of a full prepolymer having a very high viscosity, rendering it difficult to process. Lowering the level of prepolymerization lead to foams that tend to close. The second drawback is that this process is very sensitive to the presence of a polyol that has an EO content (generally as tipped) from 10 to 20 % by weight. This polyol happens to be used in almost all kinds of foams and thus the equipment will always contain some of this polyol. Carrying out the process of this patent thus requires to clean the equipment very thoroughly before use.

US-P-5594097 to Chaffanjon discloses specific polyols, having the structure PO-PO/EO-EO. One example makes use of a mixture of this polyol with an EO-rich polyol; the resulting foam is said to be of poor quality.

WO 94/24183 discloses the preparation of flexible polyurethane foams using a polyisocyanate containing at least 85% by weight of 4,4'-MDI and a polyol composition comprising 25 to less than 50% by weight of oxyethylene groups; prepolymers may be used as well. Only up to 50% by weight of other polyols may be used.

US-P-5677361 to Treboux et al. discloses a process for making high resilience foams comprising reacting an isocyanate with a specific polyol composition, which comprises an EO-rich polyol, a classical polyol

15

20

25

30

comprising tipped EO, and a polymer polyol. This process however uses the EO-rich polyol in minor amounts.

The instant invention aims at solving these problems and at providing a process which is not sensitive to POrich polyols (EO contents below 20%) and which is easy to carry out, and at providing reaction systems which are easily processable. The instant invention also aims at providing foams with high resilience and low compression set values, low hysteresis loss (lower than 20%), and with latex-like feel.

The invention thus provides a prepolymer process for preparing a high resilience flexible polyurethane foam at an NCO index of 70-120, preferably 70-110, by reacting:

- a) a polyisocyanate composition comprising at least 80% by weight of 4,4'-MDI;
- b1) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, where the EO is present as tipped EO and/or random EO, the total EO content being at least 50 % by weight;
- b2) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, where the EO is present as tipped EO and/or random EO, the total EO content being between 20-50 % by weight, having a primary hydroxy content of at least 50%;
 - b3) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxyl functionality of 2-6, having a primary hydroxy content of at least 50%, and having an EO content of from 10 to 20% by weight;

these polyols b1, b2 and b3 being reacted according to the following proportions, based on the combined weights of b1, b2 and b3, b1: 40-90 wt %, b2: 5-60 wt %, b3: 0-20 wt %;

- c) water; and
- additives and auxiliaries known per se;
- and where the polyisocyanate prepolymer has an NCO value below 20% by weight.

The invention also relates to a reaction system comprising A) a polyisocyanate prepolymer obtained by reacting the polyisocyanate with part of the polyol composition of the invention, and B) an isocyanate-reactive component comprising the remainder of the polyol composition of the invention and water.

In the context of the present invention the following terms, if and whenever they are used, have the following meaning:

1) isocyanate index or NCO index:

the ratio of NCO-groups over isocyanate-reactive hydrogen atoms present in a formulation, given as a percentage:

[NCO] x 100 (%)

15

20

25

30

35

10

5

[active hydrogen]

In other words the NCO-index expresses the percentage of isocyanate actually used in a formulation with respect to the amount of isocyanate theoretically required for reacting with the amount of isocyanate-reactive hydrogen used in a formulation.

It should be observed that the isocyanate index as used herein is considered from the point of view of the isocyanate process involving the foaming ingredient and the isocyanate-reactive ingredients. Any isocyanate groups consumed in a preliminary step to (including produce modified polyisocyanates isocyanate-derivatives referred to in the art as quasi or semi-prepolymers and prepolymers) or any active hydrogens reacted with isocyanate to produce modified polyols or polyamines, are not taken into account in the calculation of the isocyanate index. Only the free isocyanate groups and the free isocyanate-reactive hydrogens (including those of the water) present at the actual foaming stage are taken into account.

2) The expression "isocyanate-reactive hydrogen atoms" as used herein for the purpose of calculating the

15

20

25

30

35

isocyanate index refers to the total of hydroxyl and amine hydrogen atoms present in the reactive compositions in the form of polyols, polyamines and/or water; this means that for the purpose of calculating the isocyanate index at the actual foaming process one hydroxyl group is considered to comprise one reactive hydrogen, one primary or secondary amine group is considered to comprise one reactive hydrogen and one water molecule is considered to comprise two active hydrogens.

- 3) Reaction system: a combination of components wherein the polyisocyanate component is kept in a container separate from the isocyanate-reactive components.
 - 4) The expression "polyurethane foam" as used herein generally refers to cellular products as obtained by reacting polyisocyanates with isocyanate-reactive hydrogen containing compounds, using foaming agents, and in particular includes cellular products obtained with water as reactive foaming agent (involving a reaction of water with isocyanate groups yielding urea linkages and carbon dioxide and producing polyurea-urethane foams).
 - term "average nominal 5) The functionality" is used herein to indicate the average functionality (number of hydroxyl groups per molecule) of the polyol composition on the assumption that this is the average functionality (number of active hydrogen atoms of the used in initiator(s) molecule) preparation although in practice it will often be somewhat less because of some terminal unsaturation. average equivalent weight of a polyol is the average molecular weight divided by this average nominal hydroxyl functionality.
 - 6) The term "average" is used to indicate an average by number.
 - 7) The term "high resilience foams" are intended to designate those foams having a resilience of

25

30

35

at least 40%, as measured according to ISO 8307.

The following way of describing polyols is used in the present application : A PO-EO polyol is a polyol having first a PO block attached to the initiator 5 followed by an EO block. A PO-PO/EO polyol is a polyol having first a PO block and then a block of randomly distributed PO and EO. A PO-PO/EO-EO polyol is a polyol having first a PO block then a block of randomly distributed PO and EO and then a block of EO. 10 polyol is a polyol having first a PO block and then an EO In the above descriptions only one tail of a polyol is described (seen from the initiator); the nominal hydroxy functionality will determine how many of such tails will be present. 15

The polyisocyanate used in the prepolymer comprises at least 80% by weight of 4,4'-MDI (methylene diphenyl isocyanate).

The remaining part may comprise isomers thereof, higher functionalities oligomers thereof, variants thereof, or any other isocyanate (such as TDI). The isomers can be 2,4'-MDI and/or 2,2'-MDI. The oligomers are known, like e.g. polymeric MDI having isocyanate functionalities above 2. Variants thereof are known carbodiimide, uretonimine, containing variants isocyanurate, urethane, alophanate, urea and/or biuret groups. Most preferred isocyanates are pure 4,4'-MDI (isomer content less than 4% by weight), mixtures of 4,4'-MDI with polymeric MDI, mixtures of 4,4'-MDI with uretonimine and/or carbodiimide and/or urethane modified MDI.

The polyisocyanate may contain dispersed urea particles and/or urethane particles prepared in a conventional way, e.g. by adding a minor amount of an amine such as isophorone diamine to the polyisocyanate.

10

15

20

25

30

35

The prepolymer is obtained by standard methods known to the skilled man. The NCO value is below 20% by weight (the value of 20% by weight is excluded), for example from 7 to 20% by weight, preferably below 15% by weight. Preferred range is 7 to 15% by weight.

Polyol bl is an EO rich polyol. It can be prepared by known methods. It comprises PO and EO, where the EO can be random, tipped, or both. Preferably the EO is random in majority. The EO content is greater than 50% by weight (over the total oxyalkylene units present).

Polyol b2 may have a structure of the type PO-PO/EO-EO or of the type PO/EO-EO or of the type PO-EO. The total EO content is from 20 to 50 % by weight (over the total oxyalkylene units present), preferably from 21 to 45 %. Polyol b2 has a primary OH content of at least 50%, preferably at least 70% based on the primary and secondary hydroxyl groups in the polyol. In the PO-PO/EO-EO type polyol, the first PO block comprises preferably from 20 to 75 % by weight of the PO units. In those polyols b2) comprising both tipped and random EO, the weight ratio tipped EO/random EO preferably is from 1:3 to 3:1. The polyol having a structure of the type PO-PO/EO-EO can notably be produced according to the teaching of the Chaffanjon et al. US-A-5594097. polyol having a structure of the type -PO/EO-EO can notably be produced according to the teaching of the Hostettler US-A-4559366. One example of a polyol b2) is one where the tipped EO content is from 10-20 % by weight.

Polyol b3 is an optional polyol. It can be prepared by known methods. It has a primary hydroxy content of at least 50%, preferably at least 70%. It can be of various structures (PO-EO, PO/EO, etc.), where the EO can be either random, tipped, or both. The EO content is from 10 to 20% by weight (over the total oxyalkylene units present). A preferred polyol is one where EO is present as tipped EO.

10

15

20

25

30

35

The nominal functionality of these polyols b1, b2 and b3 is comprised between 2 and 6, preferably between 2 and 4.

The average equivalent weight of these polyols b1, b2 and b3 is generally comprised between 1000 and 4000, preferably 1500 and 3500.

The polyols are caused to react in the process according to the following proportions, expressed on the basis of the combined weights of the polyols:

b1: 40-90 %, preferably 50-80 % by weight

b2 : 5-60 %, preferably 10-40 % by weight

b3: 0-20 %, preferably 0-10% by weight.

Each component bl, b2 and b3 may be comprised of mixtures.

Dispersed material can also be present. This is known as polymer-modified polyol, and comprise e.g. SAN or PIPA (Poly Isocyanate Poly Addition), or PHD (polyurea suspension).

The polymer-modified polyols which are particularly interesting in accordance with the invention are products obtained by in situ polymerization of styrene and/or acrylonitrile in poly(oxyethylene/oxypropylene)polyols and products obtained by in situ reaction between a polyisocyanate and an amino- or hydroxy-functional compound (such as triethanolamine) in a poly(oxyethylene/oxypropylene)polyol. The solids content (based on the total polyol weight b1+b2+b3) can vary within broad limits, e.g. from 5 to 50 %. Particle sizes of the dispersed polymer of less than 50 microns are preferred. Mixtures can be used as well.

Water is used as the blowing agent. Carbon dioxide may be added if needed. It is appropriate to use from 1.0 to 15%, preferably from 2 to 6%, by weight of water based on the weight of the total polyol component (prereacted and not prereacted, i.e. the total starting polyol or total starting isocyanate-reactive compounds),

WO 01/32735

5

10

15

20

25

30

35

where the water can optionally be used in conjunction with carbon dioxide.

Other conventional ingredients (additives and/or auxiliaries) may be used in making the polyurethanes. These include catalysts, for example, tertiary amines and organic tin compounds, surfactants, cross linking or chain extending agents, for example, low molecular weight compounds such as diols, triols (having a molecular weight below the one of b3) and diamines, flame proofing agents, for example, halogenated alkyl phosphates, fillers and pigments. Foam stabilizers, for example polysiloxane-polyalkylene oxide block copolymers, may be used to stabilize or regulate the cells of the foam.

The amount of these minor ingredients used will depend on the nature of the product required and may be varied within limits well known to a polyurethane foam technologist.

These components, notably the polyols b1, b2 and b3 can be added in any order, provided a prepolymer is used according to the NCO values indicated above. The polyols can be added one after the other, or part by part, in any order (for example, part of b1, then the remainder of b1 together with the all of b2+b3 or all of b2 then all of b1 then all of b3).

In one embodiment, the prepolymer is obtained with all the polyol bl, and no polyol b2 and b3, which are thus added in the resin side.

The components of the polyurethane forming reaction mixture may be mixed together in any convenient manner, for example the individual components may be pre-blended so as to reduce the number of component streams requiring to be brought together in the final mixing step. It is often convenient to have a two-stream system whereby one stream comprises the polyisocyanate prepolymer and the second stream comprises all the other components of the reaction mixture.

WO 01/32735

5

10

15

20

25

30

The flexible foams may be made according to the moulding or the slabstock technique. The foams may be preferably prepared by known moulding processes, hot or cold. The foams may be used in the furniture and automotive industries in seating, cushioning and mattresses.

The flexible foams thus obtained have a free rise density comprised between, e.g. 30 and 80 kg/m^3 , preferably 35 and 65 kg/m³. These foams show a resilience higher than about 45 %, preferably higher than about 50 %.

The invention also provides a reaction system that will enable the foam producer to make the desired foam by reacting the polyisocyanate and polyol compositions. This approach provides a relatively simple way to the customer to make a product from often a variety of chemicals by combining two compositions under appropriate conditions. The polyisocyanate component is the prepolymer, while the isocyanate-reactive component comprises the remainder of the polyol. Under this embodiment, any combination is foreseen. It covers notably the following possibilities: Part of b1+b2+b3 in the prepolymer, then the remainder of b1+b2+b3 in the isocyanate-reactive component; Part of b1+b2 but no b3, then the remainder of b1+b2 and all b3; Part of b1+b3 but no b2, then the remainder of b1+b3 and all b2; all of b1, then the all of b2+b3; all of b2, then the all of b1+b3; Part of b1, then the remainder of b1 together with the all of b2+b3; Part of b2, then the remainder of b2 together with the all of b1+b3. In one embodiment, the reaction system comprises A) a prepolymer obtained with all the bl polyol and B) the remainder of the polyol and the other reactants.

The following examples illustrate the invention without limiting it.

35 Unless otherwise indicated, all parts are given by weight.

Gl	os	sa	ry	٠,

(all functionalities are nominal functionalities)

PO/EO, with EO as random. EO content is 75 % by weight. Average equivalent weight is 1336. Functionality is 3, OH value is 42 mg KOH/g.

PO-PO/EO-EO, total EO content is 28,6 % by weight. Tip EO content is 15 % by weight.

Average equivalent weight is 2004.

Functionality is 3, OH value is 28 mg

KOH/g. Primary OH content is 87%.

PO-PO/EO-EO, total EO content is 21 % by weight. Tip EO content is 15 % by weight.

Average equivalent weight is 2004.

Functionality is 3, OH value is 28 mg

KOH/g. Primary OH content is 83%.

POlyol D
PO/EO-EO, total EO content is 29.6 % by
weight. Tip EO content is 15 % by weight.
Average equivalent weight is 2004.
Functionality is 3, OH value is 28 mg
KOH/q. Primary OH content is 84%.

PO-EO, with EO as tipped. EO content is
15 % by weight. Average equivalent weight
is 2004. Functionality is 3, OH value is 28
mg KOH/g. Primary OH content is 81%.

Polymer polyol, with 20% by weight of dispersed urethane solids from triethanolamine and uretonimine modified MDI in polyol E.

Polyol G PEG having an average equivalent weight of
442. Functionality is 3.

Isocyanate A Prepolymer based on pure 4,4'-MDI (functionality is 2) and polyol A. Functionality is 2.13. NCO value is 7.9% by weight.

27.2% by weight uretonimine-modified MDI. Functionality is 2.12. NCO value is 29.3% by weight.

B4113 Surfactant from Goldschmidt
Niax Al Catalyst from Union Carbide
D33LV Catalyst from Air Products

Foams are produced according to the following scheme. Polyols, catalysts, surfactant and water are mixed prior to the addition of isocyanates. Polyol blends and isocyanates are mixed at 20°C during 8 seconds before foaming. Free-rise foams are made in plastic buckets of 2.5 1 to 10 1.

The properties of the foam are determined according to the following methods and standards:

10 Nature of cells :

5 ·

Recession (%)

FRD (Free Rise Density)

Foaming

ISO 845

Cream Time (s)

15 End Of Rise (s)

Compression hardness

CLD 40% (kPa) ISO 3386-1

Hysteresis Loss (%)

Compression set (thickness) ISO 1856

20 Dry 75 % (%)

Humid 75 % (%)

Resilience (%)
ISO 8307
ISO 8067

Tear strength (N/m)

Max

25 Tensile strength(kPa) ISO 1798

Elongation (%)

The results are summarized in the following tables.

			,			,		. –			,	_			_
	12		35						2.5	0.5	0.1	9.0	100	15	97
	11		23						2.2	0.5	0.1	9.0	100	10	98
	10				35				2.5	0.5	0.1	9.0	100.	15	97
	6	111	12						2.2	0.5		0.5	100	10	86
	8	9	17						2.2	0.5		0.5	100	10	86
oles	7		30			N			2.5	0.5	0.1	9.0	100	15	97
Examples	9		35						2.5	0.5	0.1	9.0	100	15	97
	5			70					3.3			0.5	100	30	86
	4		40						4	0.5		0.5	100	25	77
	3		35					5	3	0.5		0.5	114	25	106
	.5.		32		-		10		2.6	1		0.3	100	25	16
	1		40						3			0.5	100	25	100
	Component	Polyol A	Polyol B	Polyol C	Polyol D	Polyol E	Polyol F	Polyol G	Water	B 4113	Niax Al	D 33 LV	Isocyanate A	Isocyanate B	NCO index

						Examples	les					
		,	,		u	9	7	α	6	10	11	12
Droport i pe	.⊣	7	ກ	4	7	2	,	,			9	3000
Proper cres	*4000	neuc	*nenc	onen*	open	oben	oben	oben	oben	oben	oben	uado
Cells	nado	open.)) (•		و	-	20	10	7
Recession %	ɔ :	⊃ <u>{</u>	> ^c	> {	4	α γ		5.4	55			
FRD (kg/cm ³)	42	28	4			2	;;;	,				
Foaming						7	7					_
Cream Time (s)						0 0	0 10	1 60	г п	o v	α	103
End Of Rise (S)						93	2	TOOT	201		2	3
Compression hardness							ر ر			0	2.4	2,3
(CI,D 40 % (kPa)	2.3	4.1	2.8	7		0.7	3.6			7	12.2	14.4
Hvateresis (%)	18.9	17.1	19.6	19.2		18.3	10.0			,		
Compression set (thick)	 									ک ک	٠,	4
Drv 75 % (%)		-			_					7	-2.3	13
Humid 75 % (%)		`						5	-	5.2.6	50 5	47 5
Resilience (%)	28	62	51	48				70	7	0:50	2	
Tear strength										151.8	135.4	156.9
Max (N/m)										60.3	43	
Tensile strength (kPa)										666	102.4	
Elongation (%)		_										

* borderline

CLAIMS

5

10

15

20

25

35

- 1. A prepolymer process for preparing a high resilience flexible polyurethane foam at an NCO index of 70-120 by reacting:
- a) a polyisocyanate composition comprising at least 80% by weight of 4,4'-MDI;
- b1) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, where the EO is present as tipped EO and/or random EO, the total EO content being at least 50 % by weight;
- b2) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, where the EO is present as tipped EO and/or random EO, the total EO content being between 20-50 % by weight, having a primary hydroxy content of at least 50%;
- b3) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxyl functionality of 2-6, having a primary hydroxy content of at least 50%, and having an EO content of from 10 to 20 % by weight;

these polyols b1, b2 and b3 being reacted according to the following proportions, based on the combined weights of b1, b2 and b3, b1 : 40-90 wt %, b2 : 5-60 wt %, b3 : 0-20 wt %;

- c) water; and
- d) additives and auxiliaries known per se; and where the polyisocyanate prepolymer has an NCO value below 20% by weight.
- 2. The process according to claim 1, wherein these polyols b1, b2 and b3 are reacted according to the following proportions, based on the combined weights of b1, b2 and b3, b1: 50-80 wt %, b2: 10-40 wt %, b3: 0-10 wt %.
 - 3. The process of claims 1-2, wherein the polyol b2) is of the -PO-PO/EO-EO type.

PCT/EP00/09731

WO 01/32735

4. The process of claims 1-2, wherein the polyol b2) is of the -PO/EO-EO type.

- 5 5. The process of claims 1-2, wherein the polyol b2) is of the -PO-EO type.
 - 6. The process of claims 1-5, wherein the functionality of the polyols b1, b2 and b3 is 2-4.
- 7. The process of claims 1-6, wherein the polyols bl to b3 further comprise dispersed particles.
- 8. The process of claims 1-7, wherein the NCO value is from 7 to 15% by weight.
- 9. A reaction system comprising A) a polyisocyanate prepolymer having an NCO value of from 7 to 20% by weight, obtained by reacting the polyisocyanate with part of the polyol composition according to claims 1-7, and B) an isocyanate-reactive component comprising the remainder of the polyol composition of claims 1-7 and water.
- 25 **10.** The reaction system of claim 9, in which the part of the polyol composition is comprised of the polyol bl).

INTERNATIONAL SEARCH REPORT

Inter Inal Application No PCT/EP 00/09731

A. CLASSI IPC 7	FICATION OF SUBJECT MATTER C08G18/48 C08G18/10		
ŧ			
	o International Patent Classification (IPC) or to both national classific	cation and IPC	
	SEARCHED cumentation searched (classification system followed by classification system followed by classific	ion symbols)	
IPC 7	CO8G	,	
Documenta	tion searched other than minimum documentation to the extent that	such documents are included in the fields se	earched
Electronic d	ata base consulted during the international search (name of data be	ase and, where practical, search terms used)
WPI Da	ta, EPO-Internal, PAJ		
C DOCUM	ENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the re	levant passages	Relevant to claim No.
Calogory			
A .	EP 0 557 792 A (BASF) 1 September 1993 (1993-09-01) page 3, line 44 -page 7, line 33 1,2,6; example 4	; claims	1
A .	WO 94 24183 A (IMPERIAL CHEMICAL INDUSTRIES) 27 October 1994 (199 page 1, line 20 -page 3, line 5; table 1		1
A	EP 0 609 982 A (IMPERIAL CHEMICA INDUSTRIES) 10 August 1994 (1994 cited in the application page 3, line 2 -page 4, line 24; 11; table 1	-08-10) example	
	·	-/	
X Furt	her documents are listed in the continuation of box C.	X Patent family members are listed	in annex.
A docume consider a docume filing of the which citatio other	ent which may throw doubts on priority claim(s) or is cited to establish the publication date of another n or other special reason (as specified) ent referring to an oral disclosure, use, exhibition or means	 'T' later document published after the interest or priority date and not in conflict with cited to understand the principle or the invention 'X' document of particular relevance; the cannot be considered novel or cannot involve an inventive step when the do 'Y' document of particular relevance; the cannot be considered to involve an in document is combined with one or materials, such combination being obvious in the art. 	the application but every underlying the claimed invention to current is taken alone claimed invention ventive step when the pre other such docu-
later ti	ent published prior to the international filling date but han the priority date claimed	'&' document member of the same patent	
Date of the	actual completion of the international search	Date of mailing of the international sea	arcu report
	3 January 2001	01/02/2001	
Name and I	mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040. Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Bourgonje, A	

INTERNATIONAL SEARCH REPORT

Inter. Inal Application No PCT/EP 00/09731

.(Continua	tion) DOCUMENTS CONSIDERED TO BE RELEVANT		
ategory °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim	No.
	EP 0 331 941 A (MOBAY) 13 September 1989 (1989-09-13) page 2, line 34 -page 4, line 4; examples	1	
\	EP 0 296 449 A (BAYER) 28 December 1988 (1988-12-28)	1	
	page 3, line 8 -page 4, line 5; example 16		
	·		
		·	

INTERNATIONAL SEARCH REPORT

..ormation on patent family members

Inter anal Application No
PCT/EP 00/09731

Patent document cited in search report	rt	Publication date		Patent family member(s)	Publication date
EP 557792	A	01-09-1993	DE	4205934 A	02-09-1993
2. 00.752	, ,		ĀŤ	140017 T	15-07-1996
			CA	2090662 A,C	28-08-1993
		•	DE	59303097 D	08-08-1996
			DK	557792 T	29-07-1996
			ES	2088603 T	16-08-1996
			US	5374667 A	20-12-1994
			US	5534185 A	09-07-1996
		· 			
WO 9424183	Α	27-10-1994	CA	2158648 A	27-10-1994
			CN	1120848 A,B	17-04-1996
			CN	1183422 A	03-06-1998
			EP	0694047 A	31-01-1996
		•	JP	8511286 T	26-11-1996
			US	5459170 A	17-10-1995
EP 609982	: <u>`</u> A	10-08-1994	AU	668784 B	16-05-1996
LI 00330L	••	20 00 1331	AU	5310294 A	04-08-1994
			CA	2113946 A	03-08-1994
		•	CN	1095386 A,B	23-11-1994
•			DE	69406319 D	27-11-1997
			DE	69406319 T	19-03-1998
			ES	2108372 T	16-12-1997
			HK	1003835 A	06-11-1998
		•		6256454 A	13-09-1994
			JP		31-08-1994
			MX	9400820 A	
•			ÜŜ	5565498 A 5594097 A	15-10-1996 14 - 01-1997
			US 		
EP 331941	A	13-09-1989	DE	3806476 A	14-09-1989
			AU	3090989 A	07-09-1989
			CA	1319786 A	29-06-1993
			DE	58904380 D	24-06-1993
			ES	2054895 T	16-08-1994
			JP	1263110 A	19-10-1989
			JP	2587290 B	05-03-1997
			US	5063253 A	05-11-1991
EP 296449	A	28-12-1988	DE	3721058 A	05-01-1989
L. 250445	••	10 11 1700	AU	604131 B	06-12-1990
			AU	1823888 A	05-01-1989
			CA	1309803 A	03-11-1992
		·	DE	3862167 D	02-05-1991
			DK	349588 A	27-12-1988
•			JP	1022915 A	25-01-1989
			US	4833176 A	23-05-1989
			us	4022110 K	25 05 1909

THIS PAGE BLANK (USPTO)